

Serial No.: 09/869,985  
Reply to Office Action of: June 10, 2003  
Atty. Docket No.: GJH-0006

### REMARKS

The Examiner will note that claim 4 has been incorporated into claim 1 and the dependent claims amended accordingly. Entry of this amendment is respectfully solicited as it will place the claims in condition to be allowed, or in the alternative, in better form for appeal.

Claims 1, 2 and 4-12 were rejected under 35 USC §103(a) as being unpatentable over Sawyer (EP 419266) in view of Olah, "Hydrocarbon Chemistry."

In the rejection, the Sawyer reference is cited as disclosing a process for producing a hydrocrackate having a relatively low sulfur and nitrogen content (page 2, lines 4-6). In page 2, lines 4-6, Sawyer describes a slurry hydrotreating process for the removal of sulfur and nitrogen compounds. There is nothing in Sawyer to indicate that slurry hydrotreating and hydrocracking are interchangeable terms.  $\Rightarrow$  hydrocracking is a form of hydrotreating

Sawyer is cited as disclosing utilizing a catalyst comprised of a bulk multimetallic comprised of at least one Group VIII non-noble metal and at least two Group VIB noble metals (page 4, lines 19-30). This cited portion of Sawyer is as follows:

"Suitable catalysts for use in the present process are well known in the art and include, but are not limited to, molybdenum (Mo) sulfides, mixtures of transition metal sulfides such as Ni, Mo, Co, Fe, W, Mn, and the like. Typical catalysts include NiMo, CoMo, or CoNiMo combinations. In general sulfides of Group VII metals are suitable. "The Periodic Table of Elements referred to herein is given in Handbook of Chemistry and Physics, published by the Chemical Rubber Publishing Company, Cleveland, Ohio, 45th Edition, 1964.) These catalyst materials can be unsupported or supported on inorganic oxides such as alumina, silica, titania, silica alumina, silica magnesia and mixtures thereof. Zeolites such as USY or acid micro supports such as aluminated CAB-O-SIL can be suitably composited with these supports. Catalysts formed in-situ from soluble precursors such as Ni and Mo naphthenate or salts of phosphomolybdic acids are suitable.

In general the catalyst material may range in diameter from 1  $\mu$  to 1/8 inch. Preferably, the catalyst particles are 1 to 400  $\mu$  in diameter so that

Serial No.: 09/869,985

Reply to Office Action of: June 10, 2003

Atty. Docket No.: GJH-0006

intra particle diffusion limitations are minimized or eliminated during hydrotreating."

In response, Sawyer teaches that the catalyst materials may be unsupported or supported. Only supported catalysts are described (page 4, lines 32 et seq.). In contrast, applicants' invention relates to bulk multimetallic catalysts which are by definition unsupported (specification, page 14, lines 24-25). Furthermore, Sawyer describes mixtures of many different metals and the only trimetallic combination disclosed is CoNiMo. This is the opposite of applicants' catalytic material as it contains two Group VIII metals and no tungsten. There is nothing in Sawyer to suggest the catalyst represented by the formula in amended claim 1 wherein the ratio of  $b:(c+d)$  is from 0.5/1 to 3/1. *not present*

The Examiner goes on to state that it would have been obvious to one having ordinary skill in the art at the time the invention was made to utilize a process wherein the bulk multimetallic catalyst is a trimetallic catalyst represented by the formula,  $(X)_b(Mo)_c(W)_dO_z$ , wherein X is a Group VIII non-noble metal, the molar ratio of  $b:(c+d)$  is 0.5/1 to 3/1 because the combination of X, W, and Mo are disclosed by the Sawyer reference and this combination can be supported on an inorganic oxide and because all the constituent components are disclosed by the reference and it would be appropriate to use them in any combination effective for hydrotreating.

First, the fact that the combination X, W, and Mo cited by the Examiner is supported on an inorganic oxide is the opposite of applicants' catalytic materials which are unsupported. Second, there is nothing in Sawyer to lead one skilled in the art to the bulk catalyst set forth in applicants' amended claim 1. There is nothing to suggest the molar ratio of  $b:(c+d)$  as being from 0.5/1 to 3/1 in a bulk (unsupported) catalyst defined in amended claim 1. Third, the present catalyst material represented by the formula in amended claim 1 is not prepared by spray drying soluble salts on a support as is customary in preparing commercial catalysts. Rather, the bulk trimetallic catalyst is prepared by a solid route (page 8, last paragraph) or by reacting the components in solution to form a precipitate (page 13, lines 10-14). The resulting bulk trimetallic

Serial No.: 09/869,985  
Reply to Office Action of: June 10, 2003  
Atty. Docket No.: GJH-0006

catalyst is a unique composition as evidenced by its X-ray diffraction pattern. There is no disclosure in Sawyer as to how to prepare an unsupported catalyst.

The Examiner next notes that the Sawyer reference discloses the use of NiO and MoO<sub>3</sub> as hydrotreating catalysts (see Sawyer at page 6, lines 15-25) and concludes that it would have been obvious to one having ordinary skill in the art at the time the invention was made wherein the bulk multimetallic catalyst is amorphous and has a unique X-ray diffraction pattern showing crystalline peaks at  $d=2.53$  Angstroms and  $d=1.70$  Angstroms because diffraction peaks represent the characteristics of the reacted metal components.

In response, the Examiner's attention is directed to page 24, lines 13-25 wherein applicants state that the present bulk catalyst particles have a different pattern from those obtained by co-mixing, and conventional hydroprocessing catalysts obtained by impregnation (as in Sawyer). The unique X-ray diffraction pattern is evidence of the non-obviousness of the present bulk trimetallic catalysts.

Finally, the Examiner concludes that it would have been obvious to one having ordinary skill in the art at the time the invention was made to utilize a process with a bulk multimetallic,  $(X)_b(Mo)_c(W)_dO_z$ , where the molar ratio of  $b:(c+d)$  is 0.5/1 to 3/1, and to utilize a process wherein the molar ratio of  $c:d$  is preferably  $>0.01/1$ , and  $z = [2b+6(c+d)]/2$  because all the constituent components are disclosed by the reference and it would be appropriate to use them in any combination effective for hydrotreating.

This conclusion of obviousness is not understood and clarification is requested. NiMo is a conventional combination effective for hydrotreating. Yet as shown in the table on page 38, the present bulk metal catalysts are far more effective for denitrogenation.

Claim 3 was rejected as unpatentable over Sawyer and Olah in view of Velenyi. Velenyi was cited as disclosing a molar ratio equivalent to the range of about 9:1 to 1:9 for Mo and W with citation to the abstract, which is set forth below:

Serial No.: 09/869,985  
Reply to Office Action of: June 10, 2003  
Atty. Docket No.: GJH-0006

### "ABSTRACT

A catalyst is disclosed which comprises a molybdenum-tungsten-containing complex represented by the formula



wherein M is selected from the group consisting of one or more metals selected from any of Groups IB, IIB, IVB, VB or VIII of the Periodic Table and/or one or more of Y, Cr, Mn, Re, B, In, Ge, Sn, Pb, Th or U, or a mixture of two or more of the metals in said group; A is at least one metal selected from the group consisting of alkali metals, alkaline earth metals, Lanthanide series metals, La, Tl, or a mixture of two or more of the metals in said group; a is a number in the range of from about 1 to about 200; b is a number in the range of from about 1 to about 200; with the proviso that either Mo or W is in excess of the other, the ratio of a:b being about 4:1 or greater, or about 1:4 or less; c is a number such that the ratio of c:(a+b) is in the range of from 0:100 to about 10:100; d is a number such that the ratio of d:(a+b) is in the range of from 0:100 to about 75:100; and e is the number of oxygens needed to fulfill the valence requirements of the other elements. A process for converting gaseous reactants comprising methane and oxygen to higher order hydrocarbons using the foregoing catalyst is also disclosed."

Applicants first note that the catalyst of Velenyi is different from applicants' catalyst as Velenyi requires an "A" component. Second, the "M" component covers Cu, Ag, Au, Zn, Cd, Hg, Ti, Zr, Hf, V, Nb, Ta, Fe, Co, Ni, Y, Cr, Mn, Re, B, In, Ge, Sn, Pb, Th, U and mixtures thereof. There is nothing to suggest the trimetallic combination of applicants' amended claim 1. Third, the ratio from Velenyi for Mo and W would range from 1:200 to 200:1. The examples from Velenyi all show a Mo:W ratio of 90:1 which is outside applicants' claimed range. Fourth, Velenyi relates to a process for converting methane to higher order hydrocarbons. In conclusion, Velenyi teaches a different catalyst with different components to achieve a different (and unrelated) process.

In the Examiner's response to arguments, the Examiner concludes that combining individual components known for their use and effectiveness in hydrotreating would have been obvious to one of ordinary skill in the art based on the teaching of Sawyer or Velenyi. In response, Velenyi is a four metal component catalyst for methane conversion to higher order hydrocarbons. It is not within the ordinary skill of the art to take the

Serial No.: 09/869,985  
Reply to Office Action of: June 10, 2003  
Atty. Docket No.: GJH-0006

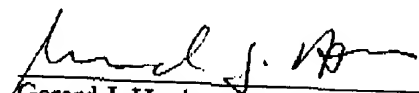
Velenyi catalyst apart, eliminate an essential metal component, select certain metals from "M" from a host of equivalent catalysts, devise a specific ratio for M to Mo+W, and then apply that to a process not even contemplated by Velenyi. Sawyer teaches conventional hydrotreating catalysts without any hint of how one might select the proper components and then prepare applicants' bulk trimetallic catalysts.

Finally, the Examiner notes that applicants fail to particularly point out how the combination of Ni, Mo and W results in unexpectedly higher activity. The Examiner's attention is directed to the comparisons in Examples 5, 9 and 20 which demonstrate the unexpected activity of the catalyst of the invention compared to a conventional Ni/Mo on alumina hydrotreating catalyst.

Based on the preceding arguments and amendments, the Examiner is requested to reconsider and withdraw all objections and rejections and pass this application to allowance. The Examiner is encouraged to contact applicants' attorney should the Examiner wish to discuss this application further.

Respectfully submitted:

Date: 8 August 2003

  
Gerard J. Hughes, Reg. No. 41,855  
Attorney for Applicants  
Telephone No.: (225) 977-4942  
Facsimile No.: (225) 977-1025

Correspondence Address:  
ExxonMobil Research and Engineering Company  
P. O. Box 900  
Annandale, New Jersey 08801-0900